Automated Image Analysis of Mineral Matter
in Raw and Supercleaned Coals

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The advent of highly sophisticated and automated microscopic tech-
·niques, together with powerful mini- and microcomputers, makes it now
possible to characterize the mineral components of coal in-situ. Com-
bined automated image analysis (AIA) and scanning electron microscopy
(SEM) allows detailed characterization of minerals in coal for size,
shape, composition, and relation to the coal matrix. For a statisti-
cally significant number of particles, both size distribution and vol-
ume fraction can be estimated and used to characterize independently
the mineral matter content. The corresponding chemical analyses for
the bulk coal samples, provided by more conventional techniques, may be
related to the AIA-SEM data for comparison.

Conventional analytical techniques such as x-ray diffraction (XRD)
and Fourier transform infrared spectroscopy (FTIR) can identify the
mineral phases present in coal and can sometimes provide an estimation
of the amount present. However, such techniques use bulk samples; they
are limited to calculating only an "average" cleaning effectiveness and
do not offer information on the size distribution of those mineral
phases identified. On the other hand, AIA-SEM permits cleaning effec-
tiveness to be evaluated with respect to both particle size and mineral
phase. Thus, problems associated with removing a particle size or
chemical class of particles can be detected and addressed. Such infor-
mation is important for any coal preparation process, especially since
it relates grinding and liberation of mineral particles to washability
tests (1).

To evaluate coal particles produced by fine grinding for washabil-
ity tests, the AIA-SEM technique provides information on the elemental
distribution among the various mineral phases. The minerals are clas-
sified using a chemistry definition file based on the relative amounts
of elements present as determined by energy-dispersive x-ray spectros-
copy.

The AIA-SEM technique has been used in Ames Laboratory to study
the effect of grinding on washability of fine coal. Several series of
coals have been characterized by this technique in the past two years,
thus demonstrating its usefulness. In this work, the AIA-SEM· technique
was applied to determine the coal mineral character before and after
cleaning.

EXPERIMENTAL

Sample Description and Preparation

The analyses were performed on two bituminous coals being tested for washability by conventional means. The coals were from the Illinois No. 6 seam, Randolph county, Illinois, and from the Pittsburgh No. 8 seam, Lewis county, West Virginia. The coals were ground to a typical power plant grind (i.e., 70-80% less than 200 mesh or 75 μm). The coals were then supercleaned by float-sink separation (using halogenated hydrocarbons) at 1.3 specific gravity to produce a very low-ash, clean coal fraction (ash content <3%). The raw and clean coal fractions were analyzed for moisture, ash, and sulfur forms by the usual ASTM procedures (see Table 1).

Table 1. ASTM analyses of raw and supercleaned coal samples for moisture, ash, forms of sulfur, and mineral matter[a].

	Illinois No. 6		Pittsburgh No. 8	
	Raw	Clean	Raw	Clean
Moisture	16.90	1.95	1.97	1.12
Ash	16.11	2.61	6.75	2.97
Total S	5.10	2.54	3.17	1.82
Pyritic S	2.37	0.22	1.35	0.03
Sulfate S	0.36	0.04	0.41	0.12
Organic S	2.36	2.27	1.42	1.67
Mineral Matter[b]	19.32	3.05	8.26	3.37

[a] Values are expressed as wt. % on a dry basis, except for moisture.
[b] Mineral matter = 1.13x ash + 0.47x (pyritic sulfur).

The raw and clean coal samples were prepared for AIA-SEM analyses by casting two grams of the sample with an epoxy resin into molds one inch in diameter. The hardened pellets were polished to reveal a cross section, with final polishing being done with 0.3 μm alumina powder. The pellets were coated with approximately 500 angstroms of carbon to render sample surfaces electrically conductive for examination in the electron microscope.

AIA-SEM Analysis

The AIA-SEM system consists of a JEOL (Japan Electron Optics Laboratory) model JSM-U3 scanning electron microscope, a LeMont Scientific B-10 image analyzer, and a Tracor Northern TN-2000 energy-dispersive x-ray analyzer. The image analyzer is a software-based system with associated electronics for SEM beam control, image amplification, and thresholding. The software base for image analysis allows the appropriate analysis algorithm to be selected for the particular sample and image conditions encountered. Particle extents are determined from the

points at which horizontal scans cross a feature, and particle outline
is reconstructed from these adjacent chords of a particle. Once the
outline has been determined, the x-ray data are collected from the
center of the particles.

Samples were analyzed in the SEM using 25 kV beam voltage, 1-2 nA
sample currents, 300x magnification, and backscattered electron imag-
ing. A point density of 1024 pixels across the screen was used to
provide ±10% accuracy on measurements as small as 1% of the field of
view. X-ray data were collected for four seconds per particle at a
typical counting rate of 1000 counts per second. Regions of interest
were set to monitor the intensities of 30 elements; however, only these
11 elements occurred with significant frequency: Na, Mg, Al, Si, P, S,
Cl, K, Ca, Ti, and Fe. Approximately 4000 particles were analyzed per
sample at a rate of 200 particles per hour.

Data Handling

Based on the relative amounts of the elements present, the parti-
cles were classified into one of nine mineral categories according to
the definitions given in Table 2. The categories derived from previ-
ously established guidelines (2,3) and included the common coal miner-
als pyrite, quartz, calcite, siderite, kaolinite, and illite. Several
other minerals were identified, but they occurred in such small amounts
that they were classified together into one common category titled
"MINORS". This category included the minerals gypsum, dolomite,

Table 2. Chemical limits for mineral phase definitions

Mineral Phase	Chemical Definition in % Range[a]	Specific Gravity
PYRITE	S 10-80; Fe 10-70	5.00
KAOLINITE	Al 15-80; Si 15-85; Al/Si 0.33-3.0	2.65
ILLITE	Al 10-50; Si 20-85; Mg 0-15; Ca 0-35; Fe 0-40; Ti 0-15	2.75
QUARTZ	Si 65-100	2.65
CALCITE	Ca 70-100	2.80
SIDERITE	Fe 70-100; Mn 0-30; Ni 0-30	5.00
MINORS (includes the following categories)		
GYPSUM	S 10-80; Ca 10-70	2.30
DOLOMITE	Mg 10-60; Ca 60-100	2.90
RUTILE	Ti 70-100	4.50
ALUMINA	Al 65-100	4.00
APATITE	P 15-40; Ca 30-100	3.20
SILICATES	Si 20-80	2.70
MISCELLANEOUS	(no restrictions, all particles accepted)	2.00

[a]Specifications may be given for the amount of other elements that
are allowed to be present. Such specifications allow minor amounts of
elements not specifically listed in the class definition to be pres-
ent, but they place an upper limit on the allowable amount.

rutile, alumina, and apatite. In addition, several other categories
were defined to accommodate particles not corresponding to any of the
above definitions. For example, a "SILICATES" category was defined to
include particles with significant silicon content, yet with the bal-
ance of the elements in such proportions that the particle would not
fit into either the quartz, kaolinite, or illite categories. A "MIS-
CELLANEOUS" category was provided to include those particles whose
composition did not allow them to fall into any of the above-mentioned
categories. Further descriptions of the instrumental, statistical,
classifying, and processing techniques will be given in another
publication being readied for submission.

The AIA procedures classified mineral particles into both size and
chemical classes. Area-equivalent diameter was used as the size param-
eter for data presentation. This measurement is the diameter of a
circle with the same area as that measured for the mineral particle.
Area-equivalent diameter was used instead of a simple length or width
measurement because the outline of mineral particles in coal is often
complex enough to render such measurements meaningless. The equivalent
diameter measurement allows particles to be classified by the area of
the particle and yet reported in terms of a linear dimension. Using
available literature values for the specific gravity of the individual
minerals, the data were then expressed as the weight fraction of the
mineral matter within a given mineral/size category. The weight frac-
tion data were then normalized using the mineral matter content to pre-
sent the mineralogical estimates on a dry coal basis. Such a presenta-
tion provides a common base for comparing the coals before and after
processing.

The data are presented in Tables 3a and 3b for the raw and clean
Illinois coal respectively, and in Tables 4a and 4b for the raw and
clean Pittsburgh coal, respectively. Percent removal of the mineral
matter calculated for each mineral phase and size category for both
coal samples is presented in Tables 3c and 4c.

<center>RESULTS AND DISCUSSION</center>

Conventional analytical data, obtained by ASTM procedures and pre-
sented in Table 1, indicate that the float-sink separation achieved an
84% and a 56% removal of ash from the Illinois and Pittsburgh coals,
respectively, with corresponding decreases in total and pyritic sulfur
of 50 and 91% and of 43 and 97%, respectively. If expressed as a re-
duction in the total mineral matter content, based on using a modified
Parr formula (4), the corresponding values are 84% for the Illinois
coal and 59% for the Pittsburgh coal.

The AIA data for the Illinois coal (Tables 3a-c) and for the
Pittsburgh coal (Tables 4a-c) are much more interesting. Results for
the raw Illinois coal show that pyrite, quartz, and two clays (kaolin-
ite and illite in approximately equal proportions) make up the bulk
(86%) of the mineral matter. The mineral phases are rather uniformly
distributed over the entire range of particle size from less than 4 μm
to more than 36 μm in diameter.

Table 3a. AIA results for Illinois No. 6 raw coal (200 x 0 mesh), expressed as weight percent of dry coal.

Particle Size

Mineral Phase	<4 μm	<7 μm	<12 μm	<21 μm	<36 μm	>36 μm	Totals
Pyrite	0.71	0.77	1.16	1.44	1.62	1.27	6.97
Kaolinite	0.57	0.67	0.65	0.38	0.36	0.20	2.83
Illite	0.32	0.38	0.42	0.42	0.45	0.47	2.47
Quartz	0.84	0.90	1.02	0.71	0.35	0.61	4.43
Siderite	0.10	0.10	0.08	0.14	0.00	0.00	0.42
Calcite	0.04	0.04	0.14	0.11	0.10	0.41	0.83
Silicates	0.23	0.16	0.17	0.10	0.03	0.19	0.89
Minors	0.02	0.03	0.05	0.01	0.04	0.00	0.15
Miscellaneous	0.05	0.06	0.11	0.03	0.07	0.00	0.32
Totals	2.87	3.12	3.79	3.36	3.02	3.15	19.32

Table 3b. AIA results for Illinois No. 6 coal floated at 1.3 specific gravity, as expressed as weight percent of dry coal.

Particle Size

Mineral Phase	<4 μm	<7 μm	<12 μm	<21 μm	<36 μm	>36 μm	Totals
Pyrite	0.19	0.28	0.42	0.21	0.07	0.00	1.17
Kaolinite	0.10	0.16	0.09	0.02	0.03	0.00	0.40
Illite	0.12	0.12	0.11	0.07	0.00	0.00	0.42
Quartz	0.16	0.19	0.18	0.09	0.01	0.00	0.63
Siderite	0.02	0.01	0.01	0.02	0.02	0.00	0.08
Calcite	0.00	0.00	0.00	0.00	0.00	0.00	0.01
Silicates	0.08	0.05	0.03	0.02	0.02	0.00	0.20
Minors	0.02	0.02	0.01	0.00	0.00	0.00	0.04
Miscellaneous	0.04	0.03	0.02	0.00	0.00	0.00	0.08
Totals	0.72	0.86	0.87	0.44	0.16	0.00	3.05

Table 3c. Use of AIA results to estimate percent removal of mineral matter from Illinois No. 6 coal. The numbers are based on values in Tables 3a and 3b before they were rounded off.

Particle Size

Mineral Phase	<4 μm	<7 μm	<12 μm	<21 μm	<36 μm	>36 μm	Totals
Pyrite	73	64	64	85	95	100	83
Kaolinite	83	77	86	94	91	100	86
Illite	61	69	74	82	100	100	83
Quartz	81	79	83	88	96	100	86
Siderite	81	85	86	89	--	100	80
Calcite	98	93	97	96	100	100	99
Silicates	67	67	81	77	45	100	77
Minors	38	51	75	100	100	100	72
Miscellaneous	30	58	83	100	100	100	75
Total	75	73	77	87	95	100	84

Table 4a. AIA results for Pittsburgh No. 8 raw coal (200 x 0 mesh), expressed as weight percent of dry coal.

Particle Size

Mineral Phase	<4 μm	<7 μm	<12 μm	<21 μm	<36 μm	>36 μm	Totals
Pyrite	0.13	0.62	0.72	0.81	1.00	1.19	4.47
Kaolinite	0.08	0.38	0.24	0.12	0.08	0.04	0.94
Illite	0.06	0.26	0.17	0.14	0.16	0.20	0.98
Quartz	0.03	0.13	0.12	0.08	0.01	0.15	0.52
Siderite	0.02	0.10	0.06	0.08	0.14	0.06	0.46
Calcite	0.00	0.02	0.01	0.02	0.00	0.00	0.05
Silicates	0.01	0.02	0.01	0.02	0.07	0.00	0.13
Minors	0.05	0.22	0.12	0.02	0.00	0.00	0.41
Miscellaneous	0.02	0.10	0.05	0.04	0.02	0.09	0.31
Totals	0.40	1.84	1.50	1.33	1.47	1.72	8.26

Table 4b. AIA results for Pittsburgh No. 8 Coal floated at 1.3 specific gravity, as expressed as weight percent of dry coal.

Particle Size

Mineral Phase	<4 μm	<7 μm	<12 μm	<21 μm	<36 μm	>36 μm	Totals
Pyrite	0.21	0.33	0.35	0.08	0.00	0.00	0.96
Kaolinite	0.28	0.45	0.21	0.04	0.00	0.00	0.98
Illite	0.24	0.35	0.11	0.01	0.00	0.00	0.70
Quartz	0.07	0.17	0.07	0.02	0.00	0.00	0.34
Siderite	0.00	0.01	0.01	0.03	0.00	0.00	0.05
Calcite	0.00	0.01	0.01	0.01	0.00	0.00	0.02
Silicates	0.02	0.02	0.01	0.00	0.00	0.00	0.05
Minors	0.02	0.03	0.01	0.04	0.00	0.00	0.10
Miscellaneous	0.06	0.08	0.03	0.00	0.00	0.00	0.17
Totals	0.91	1.43	0.80	0.22	0.00	0.00	3.37

Table 4c. Use of AIA results to estimate percent removal of mineral matter from Pittsburgh No. 8 coal. The numbers are based on values in Tables 4a and 4b before they were rounded off.

Particle Size

Mineral Phase	<4 μm	<7 μm	<12 μm	<21 μm	<36 μm	>36 μm	Totals
Pyrite	--	46	51	89	100	100	78
Kaolinite	--	--	12	64	100	100	--
Illite	--	--	37	96	100	100	28
Quartz	--	--	39	75	100	100	35
Siderite	76	95	80	63	100	100	88
Calcite	80	62	41	69	100	100	60
Silicates	--	--	32	100	100	100	62
Minors	57	86	89	--	100	100	75
Miscellaneous	--	17	42	100	100	100	45
Total	--	22	46	83	100	100	59

In the supercleaned Illinois coal, the levels of almost all miner-
als have been reduced by 60-85% somewhat uniformly for most of the
particle size ranges up to 21 μm. Above that size, the removal in-
creases sharply, with nearly complete removal for the larger sizes.
The 21 μm cut-off seems reasonable for pyrite when the following calcu-
lation is considered. A 21 μm pyrite phase (specific gravity of 5.0)
embedded in a 200 mesh (i.e. 75 μm) coal particle (specific gravity of
1.25) accounts for only 2.2% of the total volume. However, because it
accounts for 8.2% of the total weight of such a particle, it increases
the specific gravity of the total assemblage to over 1.33; it is thus
rejected in the float-sink separation. A 12 μm pyrite phase, however,
accounts for about 1% of the volume of the 74-μm assemblage, while the
specific gravity is 1.29; thus it is carried along with the float frac-
tion. Of course, since the size consist is 200 mesh x 0, there are
many small pyrite particles completely liberated or attached to smaller
sized coal particles; however, the cut-off at 21 μm is still remarka-
bly sharp. Furthermore, since the specific gravities for the other
minerals are lower than for pyrite, similar calculations for the other
minerals should yield a higher value for the particle size at the cut-
off point.

When the overall removal of 84% of the total mineral matter is
broken down by particle size, it shows an expected trend of increasing
with increasing particle size. The cleaning effectiveness ranges from
about 75% for the smaller particles (4-12 μm) to 87% at 21 μm and to
100% for particles larger than 36 μm.

The AIA results for the Pittsburgh No. 8 coal (Tables 4a-c) show a
distinctly different character of this coal. The mineral matter in the
raw sample is more than 50% pyrite. Although the total amount of clay
plus quartz is only one-fourth that of the Illinois No. 6 coal, the
clay-to-quartz ratio is much higher. In addition, a substantially
larger fraction of the mineral content is present in the larger size
ranges. This coal exhibits a much sharper cut-off in the effectiveness
of cleaning as a function of particle size. The superclean coal frac-
tion contains no particles larger than 21 μm in diameter. The smaller
size fractions show only small amounts of mineral matter removed. For
the two smallest sizes, even a slight enrichment can be seen for sever-
al mineral phases. This apparent enrichment is partially the result of
a mathematical anomaly of the normalization process and partially due
to the decrease in the total weight by the removal of other mineral
matter from the larger fractions. In any case, the absolute mineral
content in these very small size fractions is so minute that the al-
lowed analytical errors can account for the discrepancy, and the over-
all differences become negligible.

Another observation, however, should be scrutinized more closely.
In both coals, the content of pyrite as determined by AIA is consis-
tently higher than that calculated from the pyritic sulfur values ob-
tained by ASTM. Since the accuracy (reproducibility between different
labs) for ASTM analyses can be within 0.30 or 0.40% (for less than or
more than 2.0% pyritic sulfur, respectively)(5), part of the discrepan-
cy could be explained by the possible analytical error. A more plausi-
ble interpretation can be given if the ASTM leachings with nitric acid

do not remove all of the pyrite. Significant residues of pyrite have been observed previously in such leached coal samples (6). Another possible explanation can reside in the specific gravity of pyrite used. Although 5.0 is the literature value for mineral-grade pyrite, past work in this laboratory established that coal-derived pyrite can have a specific gravity significantly lower, ranging from about 3.60 for hand-picked samples to 4.25-4.50 for samples which have been extensively cleaned with hot hydrochloric acid (7). Although the absolute values for the pyrite content are divergent, the relative amounts removed (on a wt. % basis) are relatively comparable for both methods of analysis. However, further work is in progress to resolve this issue.

CONCLUSIONS

Automated image analysis used in conjunction with scanning electron microscopy and energy-dispersive x-ray analysis has been shown to be an effective tool to characterize in-situ the mineral matter in raw and cleaned coal. Both mineral phase analysis and particle size distribution were obtained for two coals (200 mesh x 0) before and after processing. For Illinois No. 6 coal, which contained mostly pyrite, quartz, kaolinite, and illite rather uniformly distributed among the various particle sizes, the cleaning effectiveness increased gradually with increasing particle size of the mineral phases. The levels of removal ranged from about 75% for the smallest particles to 100% for particles larger than 36 μm. For the Pittsburgh coal, more than half of the mineral matter was pyrite, and the pyrite was relatively coarse. The other minerals were smaller in size. During cleaning of the Pittsburgh coal, most of the large-sized mineral matter was removed, while the finer-sized mineral matter was relatively untouched. The cut-off size was approximately 21 μm.

ACKNOWLEDGEMENT

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